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13. ABSTRACT (Maximum 200 words)
The research program focused on the development and application of advanced coherent optical interactions in semiconductor and semiconductor heterostructures. The experimental results were extensive and resulted in considerable advancement of the field. Included in the data was the first observation of the coherent nonlinear optical response of a single quantum dot exciton. Based on this result, we proceeded to demonstrate the first coherent optical control of a single quantum dot exciton and the basic features of quantum wave function engineering in these systems. Our measurements also included the first demonstration of two-electron entanglement in these systems. With support with an ARO-DURIP program, we have succeeded in building the first low temperature near field optical scanning microscope based on probing the coherent nonlinear optical response. The data show evidence that we have been able to map the wave function of a localized exciton in a quantum dot.

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FINAL REPORT

Nonlinear Optical Studies of Semiconductor Heterostructures

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12. Gang Shen, Nicolas Bonadeo, Lu Sham, Dan Gammon, Duncan Steel, "Exciton-exciton correlations in a single quantum dot: Two electron entanglement" in preparation.

13. J. R. Guest, D. Gammon, D.G. Steel, "Near Field Imaging of the Coherent Nonlinear Optical Response: A mapping of the localized exciton wave function in GaAs quantum dots" in preparation.

INVITED PAPERS

1. D.G. Steel "Effects of Optical Induced Coherences in Semiconductors: An Overview", OSA/ILS'95.
2. D.G. Steel, "Application of Coherent Nonlinear Laser Spectroscopy to the Study of Disorder," Workshop on Disorder, Marburg, Germany (1995).
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Photon Coherently Excited Biexciton," QELS'95, OSA Technical Digest **16**, p246 (1995).
5. N. H. Bonadeo, R Merlin and D. G. Steel, "Ultrafast Electric Field Polarization Evolution in the Excitonic Free Polarization decay of GaAs," QELS'96.
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7. C. Schaefer and D. G. Steel, "The effects of momentum scattering on exciton motion: Observation of non-diffusive transport," QELS'96.
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15. J.C. Kim, J. Erland, D.G. Steel, and D Gammon, "Exciton Dynamics in Disordered Energy Landscape," APS March Meeting (1998).
16. J. Erland, N.H. Bonadeo, D. Gammon, and D.G. Steel, "Coherent Control of a Quantum Dot Exciton Wave Function", EQEC 98, 14-18 September 1998, Glasgow, Scotland, Post Deadline Paper EPD2.5.
17. J. R. Guest, T. H. Stievator, A. S. Lenihan, Gang Chen, D. Gammon,, D. S. Katzer, D. Park, D. G. Steel, "Nano-Optics: Imaging the Resonant Nonlinear Response of Individual Localized Excitons," QELS'99 (1999).
18. N. H. Bonadeo, Gang Chen D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, "Single Quantum Dot States: Energy Relaxation and Coupling," QELS'99 (1999).
19. D. Gammon, S.W. Brown, T.A. Kennedy, E.S. Snow, Gang Chen, N.H. Bonadeo, D.G. Steel, "Magneto-optical spectroscopy of single quantum dots: electron and nuclear spin," APS March Meeting (1999).

Educational Activity

A number of students participated in the program as evidenced in the above publications. Three of the students have since graduated with a Ph.D and one is now a faculty member in a tenure track position. The others have taken postdoctoral positions including one at AT&T. Two other students will be graduating within the next 18 months, including the student who has developed the low temperature

nonlinear optical NSOM. Three new students have just joined the group and will be involved in the new program.

BRIEF OUTLINE OF RESEARCH FINDINGS:

All of the research findings presented in this report have been reported in the annual reports. However, for completeness, we summarize some of the most exciting results.

This work focuses on developing and applying the necessary methodology to study electronic and optical properties of systems characterized by nanoscopic structure. The work includes critical fundamental studies in high dimensional structures to clarify fundamental light-semiconductor interactions problems as well as establishing the first measurements of the coherent nonlinear optical response and coherent control on single quantum dots. The work also includes development of the first low temperature near field scanning optical microscope (NSOM) for nonlinear optical spectroscopy.

During this program, the following major developments were achieved:

Measurement of the ultrafast two-photon induced coherent exciton oscillation and the exciton Raman coherence demonstrated the important role of exciton-exciton correlations.

The data shows clear evidence of the presence of exciton-exciton interactions leading to correlations. The measurements demonstrate the existence of Raman coherence between the light-hole and heavy-hole excitons as observed through detection of the time evolution of the relative phase between the two states of the system. In Fig. 1a, simplified 2-electron model is provided which shows the result of the perturbation solution of the density matrix equations. Only in the presence of exciton-exciton interactions are the beats observed in the measurement as seen in the model prediction of Fig. 1b. Figure 2 shows the experimental observation confirming the prediction.

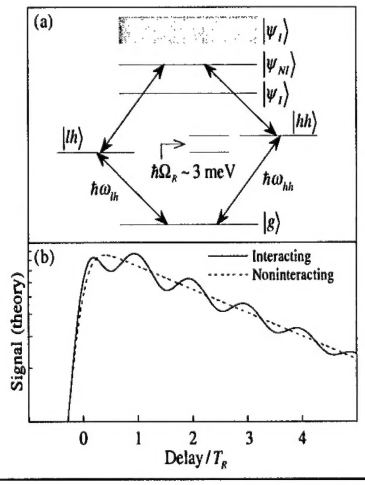

Figure 1. The *lh-hh* interaction produces bound or scattering states $|\psi_I\rangle$ (a) and produces Raman quantum beats (b: solid), which would be absent for noninteracting excitons (b: dashed). Energy differences have been greatly exaggerated for clarity. The Raman period and frequency are related by $T_R\Omega_R = 2\pi$.

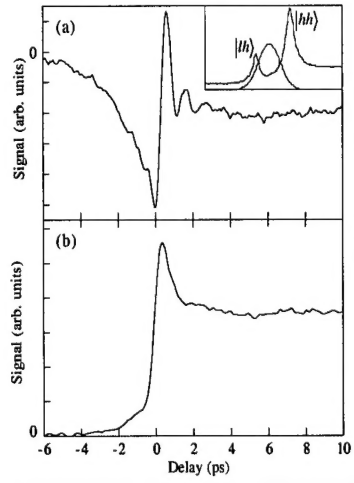

Figure 2. (a). Raman coherence oscillations appear in homodyne-detected FWM for the tuning shown in the inset . The pulse bandwidth is 2 meV, and the total excitation density is 9×10^{15} cm^{-3}. (b). For a slightly different tuning, the beats are suppressed. (The overall sign of the DT response changes from (a) to (b), reflecting the sign change in EID as the laser is tuned closer to the lh exciton resonance.)

In collaboration with D. Gammon's group at the Naval Research Laboratory, we reported the coherent nonlinear optical interaction of a single isolated quantum dot exciton. The measurements provided data on the relaxation and dephasing rates of the dot as well as demonstrating the presence of inter-dot coupling.

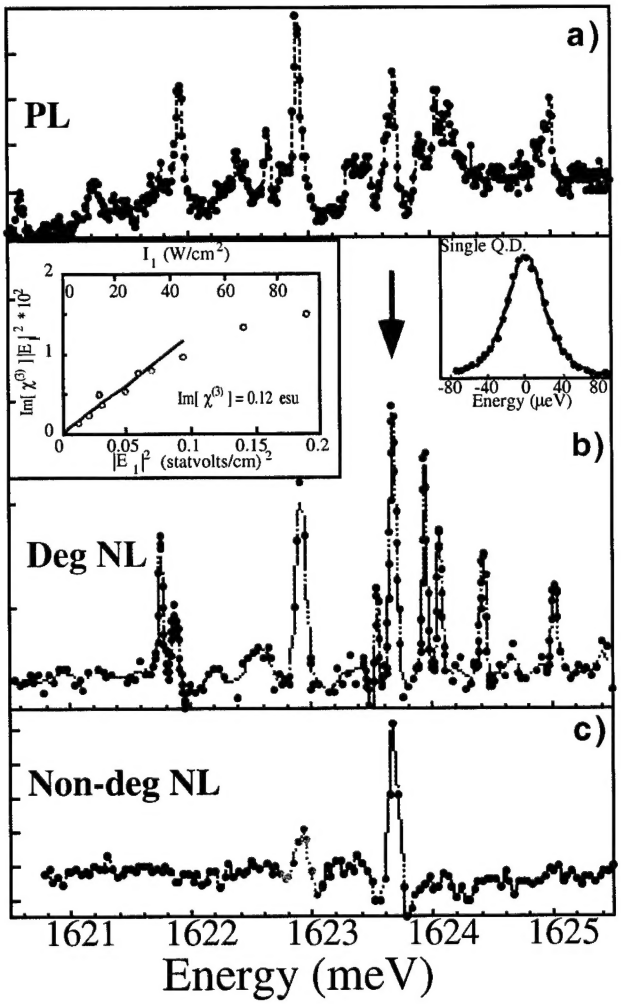

Fig. 3 (a) The PL spectrum through a 0.5 μm aperture. (b) The fully degenerate nonlinear spectrum (i. e. $\omega_1 = \omega_2$) through the same aperture. Many of the sharp lines corresponding to single QD resonances appear in both spectra, however, other resonances are visible in just one spectra demonstrating the complementary character of the probing techniques. (c) The nonlinear response as a function of ω_2 when ω_1 remains fixed at the position indicated by the arrow. (Right inset) A Lorentzian squared fit to a high resolution degenerate nonlinear response. (Left inset) The power dependence of the signal, showing a linear behavior as expected for the third order nonlinear response at low power. Saturation occurs at higher power.

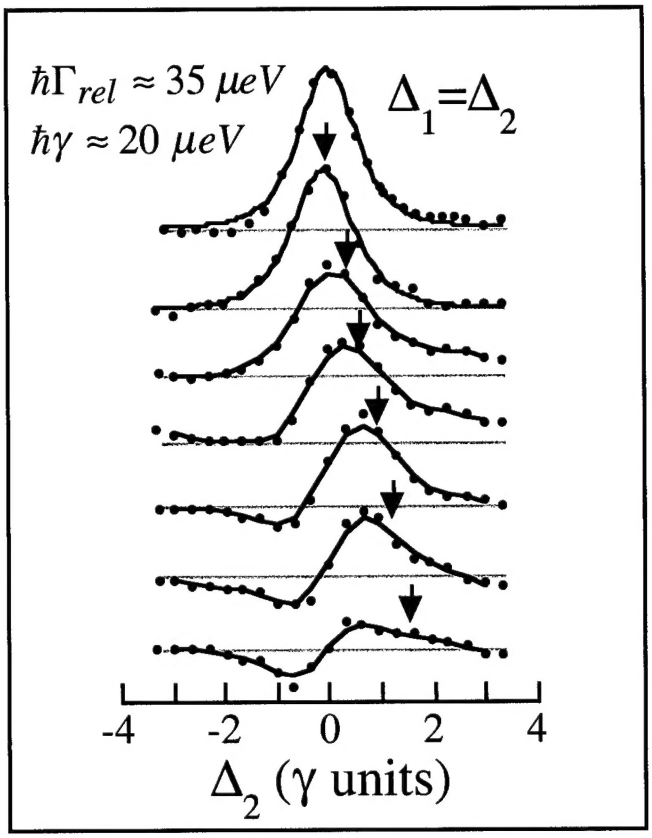

Figure 4 High resolution spectrum of the degenerate (upper curve) and nearly degenerate nonlinear response of a single QD resonance as a function of $\Delta_2 = \omega_2 - \omega_0$ for fixed detunings of $\Delta_1 = \omega_1 - \omega_0$ (located by the arrow). The data shows a clear tracking of the peak of the response, though lagging, with the position of ω_1. (Solid lines are a guide to the eye.)

In this new result, we reported the first coherent nonlinear laser spectroscopy measurements on a single quantum dot exciton. The measurements were based on degenerate and nearly-degenerate four-wave mixing measured through phase sensitive homodyne detection using high resolution spectroscopy techniques developed by us on atomic systems. The measurements in Fig. 3 compare the coherent

nonlinear optical response to PL. The bottom curve in Fig. 3 shows the nearly degenerate response demonstrating the response is fully resonant. The weaker resonances on the lower energy side demonstrate the presence of inter-dot relaxation. Figure 4 is a high-resolution spectrum of the nearly-degenerate response for an isolated quantum dot exciton as a function of pump detuning from resonance. Comparison with a model for the coherent nonlinear response shows that this data demonstrates the absence of extra-dephasing in these dots revealing an unexpected robustness in their quantum coherence against dephasing processes. The data also shows the onset of two-beam coupling.

In an extension of this collaboration with D. Gammon (Professor Lu Sham at UC-SD is also collaborating with us on this problem), we have demonstrated the role of exciton-exciton correlation in a single quantum dot showing the existence of optically induced two-electron entanglement.

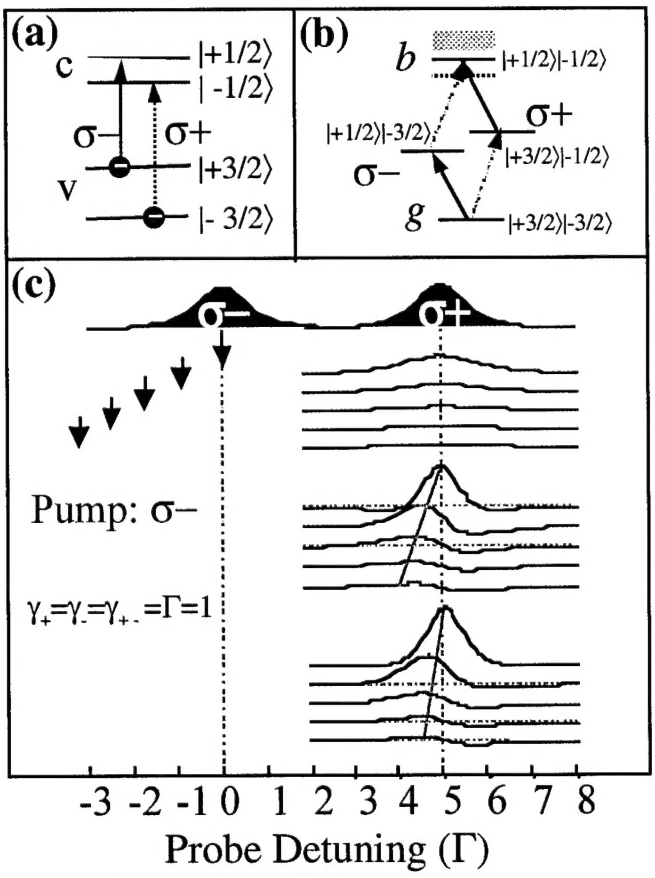

FIG. 5. (a) Band diagram for heavy-hole exciton transitions. (b) Four-level model for incorporating the four-particle Coulomb correlation into the problem. (c) Theoretical prediction for the non-degenerate experiment, assuming that the contribution from the levels beyond single-exciton levels can be neglected due to the Coulomb interaction. Curves in (A) show the incoherent contribution from the ground state depletion. Curves in (B) show the coherent contribution from the second order Zeeman coherence. Curves in (C) are the superposition of both contributions. In the absence of correlation, no signal at all would be observed.

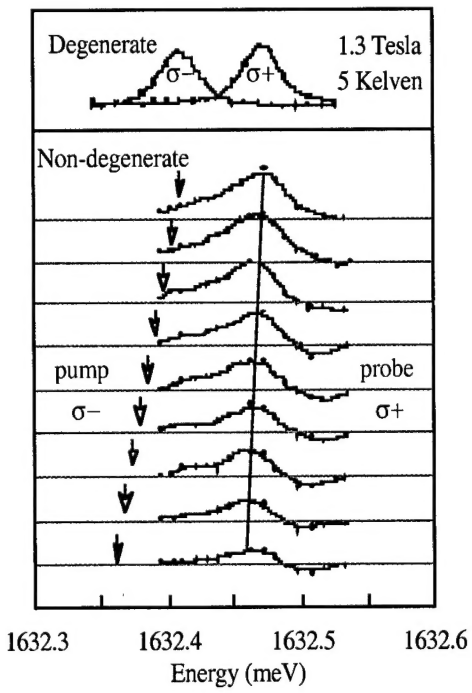

FIG. 6. Non-degenerate coherent nonlinear response. The pump is placed at the σ- (lower) state with σ- polarization. The probe is scanned across the σ+ state (upper) with σ+ polarization. The result shows interference lineshape identifying the Coulomb correlation and the contribution from Zeeman coherence (see text for details).

In the quantum dot system examined above, the correct minimal description is that of two electrons described by a basis state formed by correctly antisymmetrized eigenfunctions. Figure 5a shows the

representation based on zone center conduction and valence band electrons. Figure 5b shows the equivalent exciton representation formed by accounting for the antisymmetrization of the wave function in the two-electron basis. The results of a simple analysis of the coherent nonlinear response based on the density matrix master equations is given in Fig. 5c where the pump field at $E_1(\omega_1)$ is tuned to the σ- transition and probe field $E_2(\omega_2)$ is tuned to the σ+ transition. In the case of no interaction between the two electrons, there is no signal in the region of the σ+ transition. In the presence of just ground state depletion effects (saturation), the upper curve shows a simple resonance located at the center of the σ+ transition which does not move with tuning of ω_1. However, the second curve shows the effect of including the Zeeman coherence between the two levels. The Zeeman coherence represents a coherent superposition of the two 2-electron states excited by the σ + and σ- optical fields. This superposition represents an optically induced entanglement of the two states. The final curve represents the expected coherent nonlinear optical response in the presence of both contributions. The experimental result is quite profound and is shown in Fig. 6. The data clearly shows the unmistakable signature of optically induced entanglement and Zeeman coherence. The results show that our understanding of this system is apparently correct and that we have now established a means to demonstrate entanglement. Our future work will focus on entanglement of two dots.

Based on the coherent nonlinear optical response of the quantum dot exciton, we made the first demonstration of coherent optical control of the exciton and demonstrated the basic aspects of wave function engineering.

Using these "solid state atoms", the laboratory has now demonstrated that it can extend the concepts of coherent control and wave function engineering developed in atomic/molecular systems and higher dimensional semiconductor structures to the limit of a single quantum system in a zero-dimensional quantum dot. Such proposals have been envisioned for implementation of various schemes for quantum computation and coherent information processing and transfer in which it is important to address and coherently control individual quantum units. The quantum dots described above were used for these experiments. Symmetry breaking by the island structure mixes the polarization leading to x- and y-states of the exciton and lifts the degeneracy (data not shown).

The excitonic wave function was manipulated and monitored on a time scale short compared to the loss of quantum coherence by controlling the optical phase of two picosecond pulses through timing and polarization. Production of the two pulses and timing control was achieved using a sub-wavelength-stable Michelson interferometer to beam-split, delay and recombine the output of a picosecond laser. The experiments concentrated on the $|E_1\rangle$ state which shows a linewidth of 17 μeV and a fine structure splitting of 60 μeV (Fig 7a, inset). We probed the state of the system by monitoring the luminescence from $|E_0\rangle$.

Figure 7a shows the luminescence intensity as a function of the delay time between the phase-locked pulses when both pulses are linearly co-polarized along the Y-axis of the crystal. The resultant quantum interferogram represents the autocorrelation function of the excitonic wavefunction corresponding to state $|E_{1Y}\rangle$. The exponential decay arises from the loss of coherence.

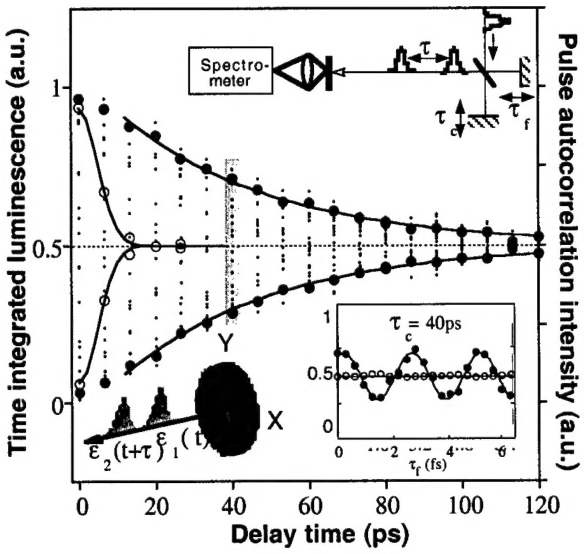

Figure 7. The amplitude of the oscillation in PL as a function of delay (large filled circles) when both pulses are co-polarized along the y-axis, thus exciting just the $|E_{1Y}\rangle$ state. This measures the autocorrelation function of the excited state wave function. The lower inset shows an expanded view around t_c=40 ps (corresponding to the shadowed region) of the small filled circles showing the oscillations in PL as a function of t_f. The large filled circles in the main figure are determined from a fit of the amplitude of oscillations as a function of t_f. The amplitude of the oscillations shows an exponential decay over long times. The auto-correlation function of the pulse is also plotted for reference (open circles). Top inset: Schematic of the experimental setup.

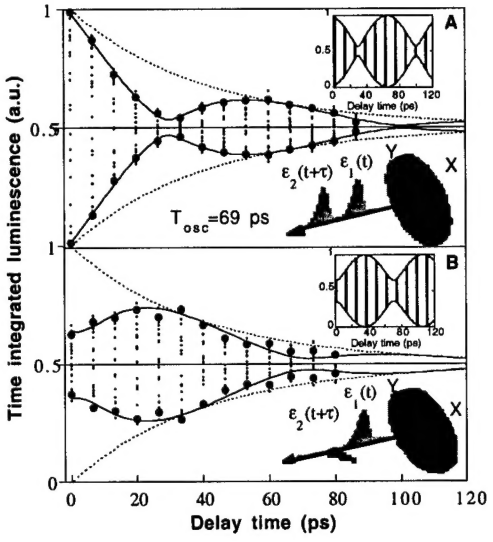

Figure 8. A) The excited state auto-correlation function of the excited state wave function, as in Fig. 2 but for both pulses co-polarized and rotated to equally excite both the $|E_{1X}\rangle$) and $|E_{1Y}\rangle$ states. The temporal evolution shows oscillations as the wave function oscillates between the two X- and Y-orthogonal states. The oscillation period corresponds to the inverse of the difference frequency between the two optical transitions. B) The cross-correlation function between two excited state wave functions generated by orthogonally polarized optical pulses. The relative phase of the two superposition of states produced by each pulse differs by π. The top inset in each figure shows the calculated oscillations in the absence of dephasing.

Even more interesting is the behavior shown in Fig. 8a in a second experiment when the polarization of the two pulses remains linearly co-polarized but oriented (~45°) to excite both the 1X and 1Y states. A non-stationary wavefunction is produced which is composed of a coherent superposition of $|E_{1X}\rangle$ and $|E_{1Y}\rangle$ states. The autocorrelation shows the wavefunction oscillating between two orthogonal states, $|E_{1X}\rangle+|E_{1Y}\rangle$ and $|E_{1X}\rangle-|E_{1Y}\rangle$. The oscillation period corresponds to the inverse of the difference frequency between the two optical transitions.

In the third experiment the polarization of the second pulse was rotated 90° relative to the first pulse of Fig 8b. The result was the creation of a superposition of the states similar to Fig 8a, but with a π shift in the quantum phase with respect to the reference wavefunction. The result is seen as a phase shift in the beating in Fig 8b. In this case the measurement reflects a cross correlation between the state of the system excited with the first pulse and the state of the system excited with the second pulse and demonstrates the ability to create a target wave function.

The results of these experiments were published in *Science*.

We developed and built a low temperature NSOM and recorded the first images of the nanoscopic coherent nonlinear optical response. The capability allowed us to map the wavefunction of localized exciton quantum dots.

A major objective of the past several years has been to develop a methodology that allows us to probe individual quantum dot structures at high density without resorting to apertures. To this end, we developed a low temperature near field scanning microscope (NSOM) designed to work based on detecting the coherent nonlinear optical response. Our first high resolution measurements are shown below. The results are quite profound and demonstrate the complexity of the localization of the dots in highly disordered systems. *The data actually provides the first mapping out of the wave function of such a system.*

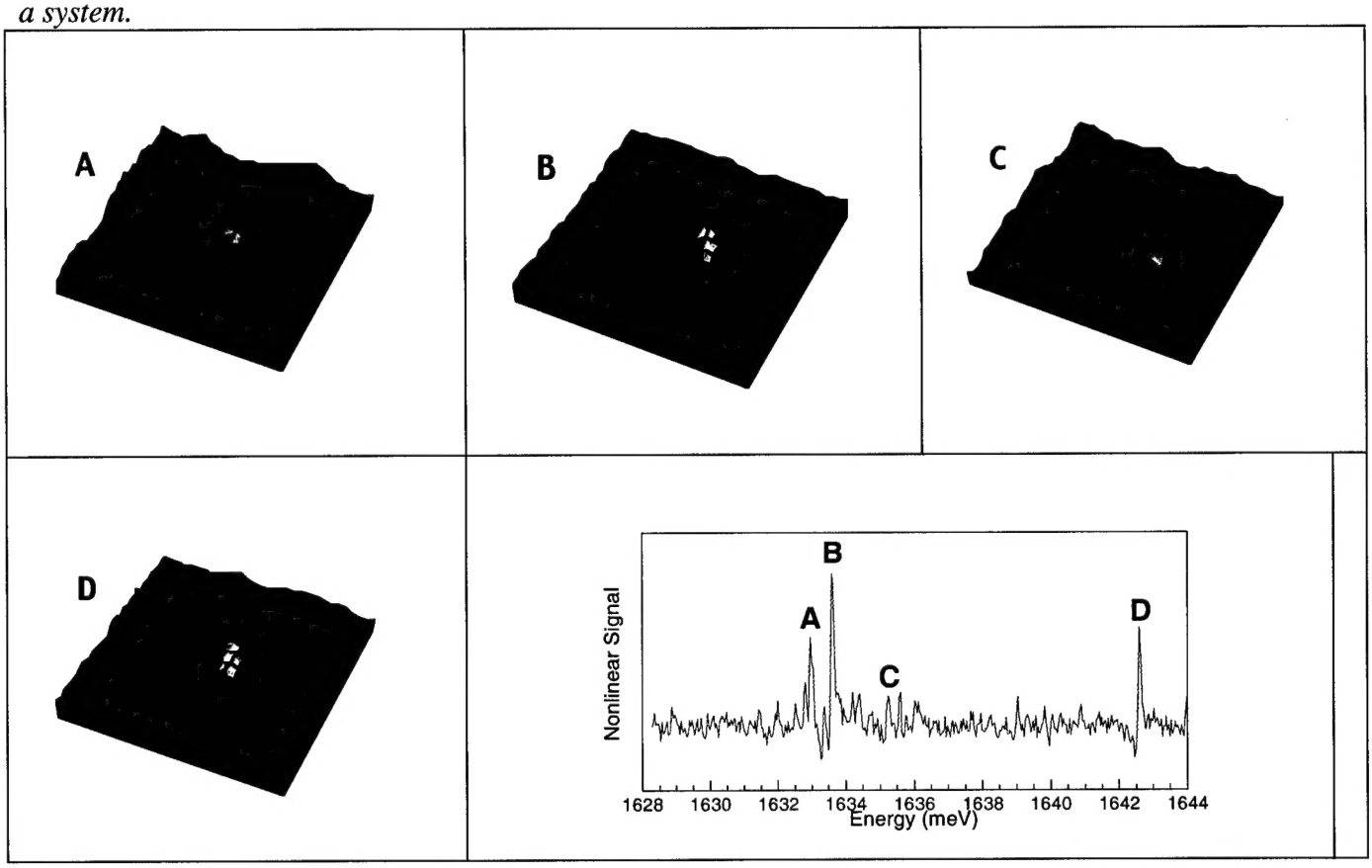

Figure 9: Degenerate nonlinear optical response images taken over 2 μm x 2 μm area at energies indicated in the spectrum (which was taken near the center of the images). Red-yellow represents positive nonlinear signal (or induced transmission), and blue represents negative nonlinear signal. Images are scaled independently. A) 1633.0 meV w/ relative signal of 2.14 B) 1633.6 meV w/ relative signal of 6.37 C) 1635.3 meV w/ relative signal of 1.55 D) 1642.7 meV w/ relative signal of 3.72. These images can be considered mappings (within the resolution of the probe) of the center-of-mass wavefunction $|\psi(x,y)|^2$ of the exciton in the disordered 2-D potential of the quantum well.